Nonequilibrium Noise as a Probe of the Kondo Effect in Mesoscopic Wires

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We study the non-equilibrium noise in mesoscopic diffusive wires hosting magnetic impurities. We find that the shot-noise to current ratio develops a peak at intermediate source-drain biases of the order of the Kondo temperature. The enhanced impurity contribution at intermediate biases is also manifested in the effective distribution. The predicted peak represents increased inelastic scattering rate at the non-equilibrium Kondo crossover.

PACS numbers: 73.23.-b, 72.15.Qm, 72.10.Fk

Recent advances in submicron physics have opened up a new window into the collective quantum physics of the electron gas away from equilibrium. One of the interesting open questions concerns the many body physics of magnetic moments coupled simultaneously to two Fermi gases with different Fermi energies. This situation has aroused great interest in the context of DC biased quantum dots [1], where a localized moment in the quantum dot is coupled to two leads at different voltage bias. Recent research have shown that magnetic moments, rather than electron-electron interactions play the dominant role in the energy relaxation of mesoscopic ("meso") copper, gold and silver wires [2, 3, 4, 5]. This discovery suggests that considerable additional insight into the nonequilibrium quantum physics of magnetic moments can be obtained from the study of mesoscopic wires.

Evidence that magnetic moments are responsible for the energy relaxation in mesowires comes from a variety of sources. The original experiments by the Saclay Group [2] showed that the broadening of the two-step distribution function in a DC biased mesowire involves a collision integral K with energy dependence $K \sim \epsilon^{-2}$. Kaminski and Glazman [3] (KG) later identified this behavior as characteristic of spin-flip scattering by magnetic impurities. Subsequent theoretical work [4, 5], and experimental confirmation [6] that magnetic fields quench the inelastic scattering [7] have essentially confirmed this hypothesis. One of the strong and as yet untested consequences of this conclusion, is that at small voltage biases, the spins inside a meso-wire should undergo the "Kondo effect", whereby the electron gas quenches the spin replacing the strong inelastic scattering by phase-coherent resonant scattering.

In this letter we consider the effects of reducing the voltage bias across a meso-wire to a value comparable and then lower than the characteristic Kondo temperature of magnetic moments in the wire. We show that in the dilute limit the impurity physics in the wire is isomorphic with that of a voltage biased quantum dot. The low frequency shot noise measures the energy relaxation in the wire, and it can be used as an alternative to the distribution function in probing the inelastic processes in

the wire. We predict that the shot-noise to current ratio will peak as the bias V is reduced to the Kondo scale T_K , and will eventually decrease back to its elastic limit as the bias is reduced even further. The predicted noise peak reflects enhanced inelastic scattering at energies of the order of $T_K[8]$.

Magnetic impurities are described by an Anderson model:

$$\mathcal{H}_{\rm imp}^{i} = \sum_{\sigma} \epsilon_{d} n_{i}^{\sigma} + U n_{i}^{\uparrow} n_{i}^{\downarrow} + W \sum_{\sigma} \left[d_{i\sigma}^{\dagger} \psi_{\sigma}(\vec{r_{i}}) + \text{H.c.} \right],$$
(1)

where $d_{i\sigma}^{\dagger}$ creates a spin- σ electron on the *i*-th impurity, n_i^{σ} is the corresponding occupation number and $\psi_{\sigma}^{\dagger}(\vec{r}_i)$ creates a conduction electron located at the same site. The impurity parameters are the orbital energy ϵ_d , the on site repulsion U and the mixing amplitude W. Another important parameter is the hybridization $\Gamma = \pi \rho W^2 \ll -\epsilon_d, \epsilon_d + U$, where ρ is the conduction band density of states. The Hamiltonian of the wire is given by $\mathcal{H} = \mathcal{H}_{\text{CB}} + \mathcal{H}_{\text{D}} + \sum \mathcal{H}_{\text{imp}}^i$, where the first two terms describe the conduction band and the disorder potential respectively.

The parameter which determines the amount of broadening in the distribution function is $n_{\rm in} = \tau_D/\tau_{\rm i}$, where $\tau_D^{-1} = D/L^2$ is the Thouless energy and $\tau_{\rm i}^{-1}$ is the energy relaxation rate due to the magnetic impurities (L and D are the wire's length and diffusion coefficient). $n_{\rm in}$ can be interpreted as the number of impurity mediated inelastic scattering events that take place during a diffusive pass across the sample. For $n_{\rm in} \ll 1$, the wire is mesoscopic and the distribution function is close to elastic distribution, while for $n_{\rm in} \gg 1$ the wire is macroscopic and the distribution function approaches a local equilibrium distribution. We shall study the mesoscopic limit and calculate the distribution and noise to first order in $n_{\rm in}$.

For a small source-drain bias $eV \ll k_B T_K$, the impurity is screened and governed by Fermi-liquid physics. The relaxation rate in this regime must increase quadratically with the bias $\tau_i^{-1} \propto V^2$. On the other hand, for the large bias regime $eV \gg k_B T_K$, the Kondo singularities are cutoff by the bias. As the bias is increased the Kondo correlations diminish and as a result the effective

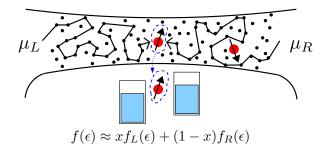


FIG. 1: For the case of $n_{\rm in} \ll 1$, each electron is scattered inelastically at most by one magnetic impurity. As a result the diffusion due to the disorder potential (dots), communicates the energy distribution in the leads to the impurities, which act as if coupled to two distinct Fermi seas.

coupling between the impurity and the conduction electrons decreases logarithmically. The energy relaxation rate in this regime should decrease like a polynomial of the effective coupling $\ln^{-1}(eV/k_BT_K)$. Based on these basic arguments the relaxation rate and $n_{\rm in}$ are maximal at some intermediate bias of the order of $T_K[9]$.

Suppose $x \in [0,1]$ is the fractional distance along the wire from the left electrode. In a wire without magnetic impurities the distribution function is elastic: $f_x^{(0)}(\epsilon) = xf_F(\epsilon - \mu_L) + (1-x)f_F(\epsilon - \mu_R)$, where $\mu_{L(R)} = \mp eV/2$, and f_F is the Fermi distribution. The corresponding shot noise relates to the current I through $S^{(0)} = 2eI/3$. In the dilute case, $n_{\rm in} \ll 1$, the distribution and the noise can be expanded in the impurity concentration c_i :

$$f = f^{(0)} + c_i f^{(1)} + \mathcal{O}(c_i^2),$$

$$S = S^{(0)} + c_i S^{(1)} + \mathcal{O}(c_i^2).$$
 (2)

The leading corrections in these expressions are the single impurity contributions. The maximum in $n_{\rm in}$ will express itself in $S^{(1)}$ and in the amplitude of $f_x^{(1)}(\epsilon)$.

The leading distribution correction is determined by making a gradient expansion of the Dyson equations of the electron Greens functions [10] which in turn yields the Boltzmann equation

$$-\frac{\partial^2 f_x^{(1)}(\epsilon)}{\partial x^2} = \tau_D I_{[f_x^{(0)}]}(\epsilon), \tag{3}$$

subject to boundary conditions $f^{(1)}|_{x=0,1}=0$. Here,

$$I_{[f_x^{(0)}]}(\epsilon) = -i \left\{ t_x^{>}(\epsilon) f_x^{(0)}(\epsilon) + t_x^{<}(\epsilon) \left[1 - f_x^{(0)}(\epsilon) \right] \right\}, \quad (4)$$

is the collision integral and $t_x^{>(<)}$ is the greater (lesser) irreducible t-matrix of a single impurity in a bath with an elastic distribution function $f_x^{(0)}(\epsilon)$.

The elastic distribution is composed of two Fermi distributions, hence the t-matrices $t_x^{<}$ and $t_x^{>}$ are exactly equivalent to t-matrices of a quantum dot which is coupled by tunneling amplitudes $\sqrt{x}W$ and $\sqrt{1-x}W$ to two electrodes at chemical potential μ_L and μ_R respectively.

This proves the equivalence of the wire problem to a quantum dot problem for $n_{\rm in} \ll 1$. Figure 1 illustrates schematically the relation between the problems.

We shall now apply a set of standard approximations to calculate the t-matrices at the small and the large bias regimes and to solve $f^{(1)}$. The noise correction $S^{(1)}$ will then be calculated through the relation [11]

$$S^{(1)} = \frac{4}{R} \int_0^1 dx \int d\epsilon \left[1 - 2f_x^{(0)}(\epsilon) \right] f_x^{(1)}(\epsilon), \quad (5)$$

where R is the resistance of the wire. Note that the non equilibrium Kondo effect is still an open problem and up to date there is no reliable description of the crossover between small and large biases. We shall employ our results in the small and large bias regimes to interpolate the physics in the crossover[9].

Small bias regime - The small voltage and low temperatures region may be treated by perturbating around the Fermi liquid fixed point. To do this, we employ an extension of Hewson's renormalized perturbation theory [12] (RPT) to weak departures from equilibrium [13]. The RPT describes the vicinity of the strong coupling fixed point by an Anderson model with renormalized parameters and counter terms to compensate for the high-energy contributions to the self-energy and vertex parts. For small departures from equilibrium we may use the same renormalized Hamiltonian but with a non-equilibrium distribution $f_x^{(0)}$ for the conduction bath.

To leading order in ϵ/k_BT_K , eV/k_BT_K and T/T_K , the generalized RPT calculation yields a self-energy with the same functional dependence on energy and voltage as a perturbative treatment of direct electron-electron scattering [14]. In particular, the energy relaxation rate is

$$\frac{1}{\tau_{\rm i}^{\rm RPT}} = \frac{\pi c_{\rm i}}{16\rho} \left(\frac{eV}{k_B T_K}\right)^2. \tag{6}$$

The solution of Eq.(3) with the collision integral calculated by the RPT gives the distribution correction to second order in ϵ/k_BT_K , eV/k_BT_K and T/T_K :

$$f_x^{(1)}(\epsilon) = -\frac{\pi L^2}{16\rho D} \sum_{\ell=-3/2}^{3/2} q_{\ell}(x) \left[\left(\frac{\pi T}{T_K} \right)^2 + \left(\frac{\epsilon - \ell eV}{k_B T_K} \right)^2 \right] \times \left[f_x^{(0)}(\epsilon) - f_F(\epsilon - \ell eV) \right]. \tag{7}$$

The spatial dependence enters through the polynomials $q_{\frac{3}{2}}(x)=\frac{1}{20}(x-x^5)+\frac{1}{6}(x^4-x^3),\ q_{\frac{1}{2}}(x)=\frac{3}{20}x^5-\frac{5}{12}x^4+\frac{1}{2}(x^3-x^2)+\frac{4}{15}x,$ and their symmetric counterparts $q_{-\ell}(x)=q_{\ell}(1-x).$ The contribution of the magnetic impurities to the noise is

$$\frac{c_{i}S^{(1)}}{S^{(0)}} = \frac{c_{i}L^{2}}{\rho D} \frac{19}{45 \cdot 7} \frac{\pi}{16} \left(\frac{eV}{k_{B}T_{K}}\right)^{2}.$$
 (8)

Large bias regime - We treat the large bias regime both numerically, using the NCA, and analytically employing

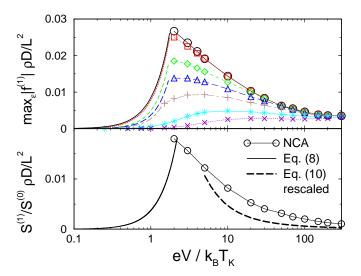


FIG. 2: Upper panel: amplitude of the distribution expansion in the middle of the wire $f_{x=1/2}^{(1)}$. The lines represent the small bias RPT expression of Eq.(7). The symbols with guide-lines show the large bias NCA results at temperatures of $T/T_K=0.001,\ 0.01,\ 0.05,\ 0.1,\ 0.2,\ 0.5$ and 1, displayed up to the intersection point with the RPT curves. Lower panel: shotnoise expansion $S^{(1)}$. The line stands for the small bias RPT expression of Eq.(8), the circles with guide-line describe the NCA results, and the thick dashed line shows the large bias asymptotic expression of Eq.(10). The NCA parameters are $\mathcal{D}/\Gamma=15,\ \mathcal{D}/\epsilon_d=-3.593$, where $2\mathcal{D}$ is the bandwidth [19].

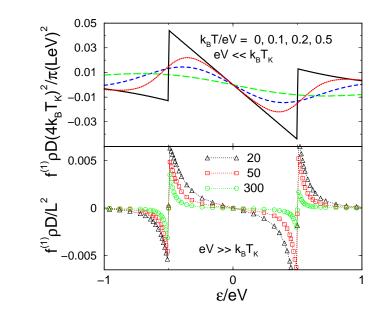


FIG. 3: The distribution expansion in the middle of the wire $f_{x=1/2}^{(1)}$. Upper panel: The small bias RPT expression of Eq.(7) at different temperatures. For a given T/V ratio the amplitude of $f^{(1)}$ is proportional to $(V/T_K)^2$ and to $L^2/\rho D$. Lower panel: NCA results at $eV/k_BT_K=20$, 50 and 300 for $T/T_K=0.001$. The parameters are the same as in Fig. 2.

a scaling argument proposed by KG. Numerically, it is convenient to calculate the impurity spectral function A_d and its effective distribution $F_d = G_d^{<}/(2\pi i A_d)$, where $G_d^{<}$ is the impurity lesser Green function. The impurity is considered within the NCA as an infinite-U Anderson impurity, coupled to a conduction bath with a non equilibrium distribution $f_x^{(0)}$. In terms of these quantities the collision integral is given by $2\rho^{-1}\Gamma A_d(F_d-f^{(0)})$. The NCA takes account of the inelastic scattering to produce a non-trivial impurity distribution function $F_d \neq f^{(0)}$ (in contrast e.g. to slave-boson mean-field) and provides a reliable description of the large bias regime. However, the NCA fails to reproduce the Fermi-liquid and cannot be fully extended to the small-bias, low-temperature regime.

Analytically, following KG, we turn now to the Kondo model rescaling the leading order perturbation theory in the exchange coupling expression by replacing $\rho J \to \ln^{-1}(eV/k_BT_K)$. This rescaling procedure describes the large bias asymptotic behavior at $eV \gg k_BT_K$. The leading amplitude of a spin mediated electron-electron inelastic process is of second order in ρJ . For the sake of simplicity the spin Green function is uniformly broadened with the Korringa rate $\tau_S^{-1} = \zeta V(\rho J)^2$, where ζ is a factor of order unity. Generally, ζ has a spatial dependence [4], however the correct physics is still captured by taking its value in the middle of the wire $\zeta = \pi/4$. To leading order in ρJ the collision integral kernel reads:

$$K(\omega) = \frac{\pi}{2\rho} \frac{3}{4} \frac{(\rho J)^4}{\omega^2 + \tau_S^{-2}},\tag{9}$$

where ω is the energy transferred between the interacting electrons. The corresponding noise correction reads

$$\frac{c_{i}S^{(1)}}{S^{(0)}} = \frac{L^{2}c_{i}}{\rho D} \frac{3\pi\eta^{2}}{80\zeta^{2}} \left[\operatorname{Re}\left\{ (1 - i\eta) \ln \frac{\eta + i}{\eta} \right\} - \frac{22}{21} \right], (10)$$

where $\eta \equiv \zeta(\rho J)^2$. Rescaling this expression we find that the impurity contribution to the noise decays at large biases like $\ln^{-4}(V/T_K)$.

Figure 2 shows the amplitude of the first order expansion of the distribution $f^{(1)}$ and the first order expansion of the shot noise $S^{(1)}$. Both quantities show a peak at intermediate biases of the order of the Kondo temperature representing the enhanced inelastic scattering rate in the crossover. The intensity of the relative contribution of the magnetic impurities to both quantities is proportional to $c_i L^2/\rho D$. The amplitude of the distribution correction has a strong temperature dependence and the peak is almost washed out at temperatures of the order of T_K . The RPT and NCA results intersect in the cross-over regime where the voltage is comparable with the Kondo temperature.

Figure 3 shows the correction of the distribution function in the middle of the wire $f_{x=1/2}^{(1)}$. This quantity measures the smearing of the two-step energy distribution by

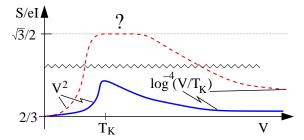


FIG. 4: The perturbative arguments hold as long as $c_1L^2/\rho D$ is small, where the curve-shape of the noise is represented schematically by the solid curve. When the impurity concentration or wire's length are increased the perturbative description breaks down at intermediate biases. Assuming that the noise has a monotonic dependence on the inelastic scattering rate, we speculate that the noise will crossover to its $\sqrt{3}eI/2$ value of local equilibrium at the intermediate biases as illustrated by the dashed curve.

inelastic impurity scattering. The correction $f_{1/2}^{(1)}$ evolves with voltage and becomes sharper, gaining an additional scale- $\eta \propto \ln^{-2}(eV/k_BT_K)$ at the large bias regime.

According to Eqs.(3) and (5) the small parameter of the c_i expansion is $\lambda = c_i \tau_D \max_{\epsilon} I_{[f^{(0)}]}$. λ acquires its maximum value at the noise peak, so we can estimate its upper bound from the RPT value at $eV = k_B T_K$:

$$\lambda(V) \le \lambda^{\text{RPT}}(k_B T_K / e) = \frac{c_i L^2}{\rho D} \frac{6\pi}{256} \approx 0.07 \frac{c_i L^2}{\rho D}.$$
 (11)

For wires in which this bound is smaller than 1, the distribution and the noise are well described by perturbation theory. The noise curve of such a wire is represented schematically by the solid curve of Fig.4. For wires in which $c_i L^2/\rho D \gtrsim 14$ the perturbative approach breaks down at intermediate biases. At very small or very large biases the energy relaxation is still small enough hence the curve will preserve its V^2 or $\ln^{-4}(eV/k_BT_K)$ asymptotics. At intermediate-small or -large biases perturbation theory breaks down in this case and one needs to solve the Boltzmann equation self-consistently like in Refs.[4, 15]. We can however speculate that in this bias regimes the wire is well described by local equilibrium and that the noise crosses over from its local equilibrium value of $\sqrt{3}eI/2$ as presented schematically by the dashed curve in Fig.4.

At present we lack a precise theoretical description of the nonequilibrium crossover regime and the detailed shape of the noise peak in this regime. In particular, it is not yet clear whether the cross-over will involve a broad maximum in the noise, reminiscent of the energy dependent inelastic scattering rate in the ground-state of the Kondo model[8], or a sharper maximum, as observed experimentally in the temperature dependent dephasing rate[16]. This issue awaits future theoretical and experimental resolution.

An interesting question for the future concerns the ex-

treme case of a heavy electron meso-wire, made up of a dense array of local moments which are quenched at zero voltage bias. Would it be possible to realize a non-equilibrium distribution in such a wire, and if so how would the non equilibrium state effect the competition between RKKY- mediated magnetism and Kondo lattice formation [17]? This is not an abstract question, for micron size filaments of the heavy electron UPt₃ are formed naturally in the melt[18].

This paper has examined the shot-noise in diffusive wires containing dilute impurities, showing that in the dilute limit the behavior of magnetic impurities inside the meso-wire is equivalent to the physics of DC biased quantum dot. The low frequency noise appears to be an ideal probe of the inelastic processes in such a non equilibrium Kondo system. We have calculated the distribution function and noise at the small and the large bias regimes. A key prediction of this paper is the appearance of a noise peak in the nonequilibrium Kondo crossover at voltages $V \sim T_K$. While many other sources of noise may exhibit a voltage dependence, this important contribution to the noise will uniquely depend on the concentration of impurities, and will also exhibit a magnetic field dependence.

Acknowledgments: We thank D. Langreth, A. Schiller, A. Silva, and M. Müller, for discussions. This work is supported by DOE grant DE-FE02-00ER45790.

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